Phase diagram of the spin-Peierls chain with local coupling

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We explore the ground-state phase diagram of a Heisenberg spin chain coupled locally to optical phonons (bond coupling), using large-scale density matrix renormalization group calculations and an extended perturbative analysis. For the quantum phase transition from the spin liquid to the dimerized phase, we find deviations from previous quantum Monte Carlo and flow equation results.

The interaction of electronic and lattice degrees of freedom in combination with reduced dimensionality can lead to a variety of interesting effects, one of which is the instability of a one-dimensional metal towards lattice distortion and the opening of a gap at the Fermi surface that was first described by Peierls[1]. A similar effect is observed in quantum spin chains, where the coupling to the lattice can cause a transition from a spin liquid with gap-less excitations to a dimerized phase with an excitation gap. Experimentally such behavior was first observed in the 1970s for organic compounds of the TTF and TCNQ family[2], but the topic regained attention after the discovery of the first inorganic spin-Peierls compound $CuGeO_3$ in 1993 by Hase et al.[3]. In this material Cu^{2+} ions form well separated spin-1/2 chains with an exchange interaction that couples to high-frequency *optical* phonons ($\omega \approx J$), and the phonon dynamics at the phase transition is governed by a central peak rather than a soft-mode behavior.[4,5] These features distinguish $CuGeO_3$ from other spin-Peierls systems and sparked the interest in a non-adiabatic modelling.

A good starting point is the study of simplified microscopic models, which can be build from three ingredients,

$$H = H_s + H_p + H_{sp}\,. \tag{1}$$

Here $H_s = \sum_i \vec{S}_i \cdot \vec{S}_{i+1}$ and $H_p = \omega \sum_i b_i^\dagger b_i$ describe a Heisenberg spin-1/2 chain and a set of harmonic (Einstein) oscillators which are coupled by an interaction term H_{sp}. For this interaction we can consider two simple forms,

$$H_{sp}^{\rm diff} = g\omega \sum_i (b_i^\dagger + b_i)(\vec{S}_i \cdot \vec{S}_{i+1} - \vec{S}_{i-1} \cdot \vec{S}_i)\,, \tag{2}$$

and

$$H_{sp}^{\rm loc} = g\omega \sum_i (b_i^\dagger + b_i)\vec{S}_i \cdot \vec{S}_{i+1}\,. \tag{3}$$

The first type of spin-phonon interaction, $H_{sp}^{\rm diff}$, has been studied with a number of methods, including perturbation theory[6,7], flow equations[8,9], exact diagonalization[7] and DMRG[10]. The latter approach identified the ground-state phase diagram, but also analytically the quantum phase transition from the gap-less to the dimerized phase is rather well understood: For finite phonon frequency ω the spin-phonon coupling g leads to effective spin interactions beyond nearest-neighbor exchange, i.e., the low energy physics is governed by a frustrated Heisenberg model. As we know from the spin model

$$H = \sum_i (\vec{S}_i \cdot \vec{S}_{i+1} + \alpha \vec{S}_i \cdot \vec{S}_{i+2})\,, \tag{4}$$

frustration can lead to dimerization if the parameter α exceeds a certain critical value ($\alpha_c = 0.241167$ in this case[11,12,13]), and similarly we obtain a finite $g_c(\omega)$.[7,8,10,14]

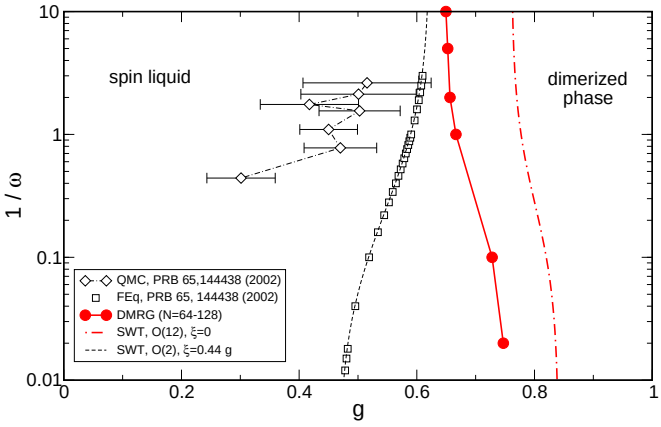

FIG. 1: (Color online) Ground-state phase diagram of the spin-Peierls chain with local coupling: QMC and flow equation (FEq) results of Raas et al.[15] compared to DMRG data and results of 12th order Schrieffer-Wolff transformation (SWT).

For the second type of spin-phonon coupling, $H_{sp}^{\rm loc}$, which applies to $CuGeO_3$, to date the precise location of the phase boundary was arguable. In previous studies[7,16] we calculated g_c using perturbation theory, a variational ansatz, as well as exact diagonalization of small systems. This was challenged by results of the flow equation method and, in a limited parameter range, by quantum Monte Carlo.[15] In this article we present unbiased results of large-scale density matrix renormalization group (DMRG) calculations, and extend our perturbation theory by several orders in g, high enough to ensure convergence. The new results are summarized in Figure 1 and compared to other approaches. More details now follow.

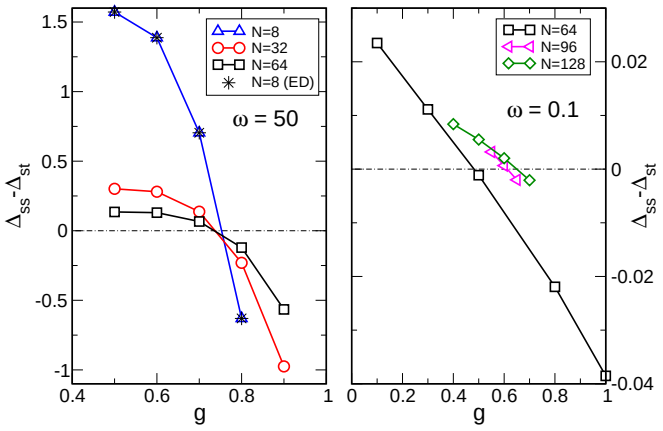

FIG. 2: (Color online) Crossing of the singlet and triplet gaps for large (right) and small (left) phonon frequency. Note the different finite-size scalings.

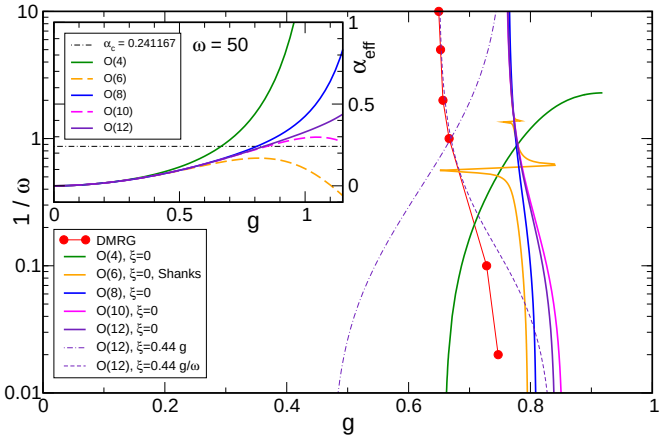

FIG. 3: (Color online) Convergence of the Schrieffer-Wolff approach with increasing expansion order $O(k)$. Main panel: phase diagram showing DMRG and expansion data for order 4–12 without and with oscillator shifts. Inset: $\alpha_{\rm eff}$ as a function of g for $\omega = 50$ and orders 4–12.

Numerical results: DMRG calculations are certainly the most precise numerical tool for studying the low energy properties of one-dimensional models, but as yet have only been performed for the spin-Peierls model with difference coupling[10] $H_{sp}^{\rm diff}$, models with acoustic phonons[17], or with XY-type spin interaction[18]. We therefore implemented the local spin-phonon interaction $H_{sp}^{\rm loc}$, using a high-performance, parallel version of the usual two-block finite-lattice algorithm with up to 1000 states per block. To detect the quantum phase transition from the gap-less to the dimerized phase we use the established criterion of the level-crossing between the first singlet and the first triplet excitation, which was derived for the frustrated spin chain[19], Eq. (4), and has successfully been applied[10,14] in the case of $H_{sp}^{\rm diff}$. For finite systems in the gap-less phase the lowest singlet excitation is above the lowest triplet, both becoming degenerate with the singlet ground state for system size $N \to \infty$. In the gapped phase, for $N \to \infty$ the lowest singlet becomes degenerate with the ground state to form the symmetry-broken dimerized state, whereas the lowest triplet maintains a finite gap. Consequently, the two excitations will cross at the critical point. Note however, that for small phonon frequency ω the relevant singlet excitation can be confused with a copy of the ground state plus an excited phonon. This frequency range therefore requires rather large N for the correct crossing to be detectable in the low energy spectrum of the spin-Peierls models.

In Figure 2 we show the level difference as a function of the spin-phonon coupling for various system sizes and two typical phonon frequencies. For $\omega \gtrsim 0.5$ there is almost no finite-size dependence of the critical coupling g_c, whereas for smaller frequencies the data scales noticeably. It is tempting to attribute this different behavior to the cross-over from the anti-adiabatic to the adiabatic regime, naïvely expected for $\omega \sim J \equiv 1$. However, as was pointed out by Citro et al.[20] using bosonization techniques, the relevant scale for the adiabatic to anti-adiabatic cross-over is given by the excitation gap, $\omega \sim \Delta$. The larger N-dependence of g_c observed in Figure 2 for $\omega = 0.1$ is therefore related to the finite-size gap still being close ω, and will disappear for $N \to \infty$. Note also, that the bosonization results support the analytical approach we present in the following, which is based on the assumption of anti-adiabaticity.

Analytical results: Already in our earlier work we suggested construction of an effective spin model for the low energy physics of the spin-Peierls problem by integrating out the phonons with a Schrieffer-Wolff transformation[21] (which removes direct spin-phonon interactions) followed by an average over the phonon vacuum. In more detail, we apply the unitary transformation $\tilde{H} = \exp(S) H \exp(-S)$, where

$$S = g \sum_i (b_i^\dagger - b_i) \vec{S}_i \cdot \vec{S}_{i+1} \,. \qquad (5)$$

Unfortunately, this transformation cannot be evaluated exactly, but needs to be approximated by an expansion in iterated commutators:

$$\tilde{H} = \sum_k [S, H]_k / k!, \qquad (6)$$

where $[S,H]_{k+1} = [S,[S,H]_k]$ and $[S,H]_0 = H$. For increasing expansion order these commutators quickly become very complicated and easily involve millions of terms. Using FORM[22,23], an algebra tool popular in high energy physics, we are now able to push the limit of the expansion to order $k = 12$, a tremendous advantage over our previous results with $k = 4$. For the last step, we decided to be more general by averaging the transformed Hamiltonian over coherent states with $b_i|\xi\rangle = \xi|\xi\rangle$ in-

TABLE I: Expansion of the effective long-ranged exchange $J_n \vec{S}_i \cdot \vec{S}_{i+n}$ in powers of the spin-phonon coupling g.

	J_0	J_1	J_2	J_3	J_4	J_5	J_6	J_7
g^0	$\xi^2\omega$	1	·	·	·	·	·	·
g^2	·	$-\frac{1}{2}$	$\frac{1}{2}$	·	·	·	·	·
$g^2\omega$	$-\frac{3}{16}$	$\frac{1}{2}$	·	·	·	·	·	·
$g^3\xi\omega$	·	$-\frac{2}{3}$	$\frac{2}{3}$	·	·	·	·	·
g^4	·	$\frac{7}{24}$	$-\frac{37}{96}$	$\frac{3}{32}$	·	·	·	·
$g^4\omega$	$\frac{3}{64}$	$-\frac{3}{8}$	$\frac{3}{16}$	·	·	·	·	·
$g^5\xi\omega$	·	$\frac{7}{15}$	$-\frac{37}{60}$	$\frac{3}{20}$	·	·	·	·
g^6	·	$-\frac{697}{5760}$	$\frac{541}{2880}$	$-\frac{29}{384}$	$\frac{5}{576}$	·	·	·
$g^6\omega$	$-\frac{1}{64}$	$\frac{29}{144}$	$-\frac{91}{576}$	$\frac{5}{192}$	·	·	·	·
$g^7\xi\omega$	·	$-\frac{697}{3360}$	$\frac{541}{1680}$	$-\frac{29}{224}$	$\frac{5}{336}$	·	·	·
g^8	·	$\frac{5119}{129024}$	$-\frac{88339}{1290240}$	$\frac{45749}{1290240}$	$-\frac{3685}{516096}$	$\frac{35}{73728}$	·	·
$g^8\omega$	$\frac{107}{24576}$	$-\frac{3611}{46080}$	$\frac{3569}{46080}$	$-\frac{683}{30720}$	$\frac{35}{18432}$	·	·	·
$g^9\xi\omega$	·	$\frac{5119}{72576}$	$-\frac{88339}{725760}$	$\frac{45749}{725760}$	$-\frac{3685}{290304}$	$\frac{35}{41472}$	·	·
g^{10}	·	$-\frac{2516207}{232243200}$	$\frac{9363217}{464486400}$	$-\frac{105569}{8601600}$	$\frac{171601}{51609600}$	$-\frac{7601}{19353600}$	$\frac{7}{409600}$	·
$g^{10}\omega$	$-\frac{18101}{17203200}$	$\frac{35017}{1433600}$	$-\frac{72439}{2580480}$	$\frac{1243}{115200}$	$-\frac{42433}{25804800}$	$\frac{7}{81920}$	·	·
$g^{11}\xi\omega$	·	$-\frac{2516207}{127733760}$	$\frac{9363217}{255467520}$	$-\frac{105569}{4730880}$	$\frac{171601}{28385280}$	$-\frac{691}{967680}$	$\frac{7}{225280}$	·
g^{12}	·	$\frac{624432139}{245248819200}$	$-\frac{820409053}{163499212800}$	$\frac{62408713}{18166579200}$	$-\frac{138813341}{122624409600}$	$\frac{12753401}{70071091200}$	$-\frac{6906257}{490497638400}$	$\frac{77}{176947200}$
$g^{12}\omega$	$\frac{69371}{309657600}$	$-\frac{441857}{68812800}$	$\frac{1701589}{206438400}$	$-\frac{7128059}{1857945600}$	$\frac{280187}{348364800}$	$-\frac{4939}{66355200}$	$\frac{77}{29491200}$	·
$g^{13}\xi\omega$	·	$\frac{624432139}{122624409600}$	$-\frac{820409053}{81749606400}$	$\frac{62408713}{9083289600}$	$-\frac{138813341}{61312204800}$	$\frac{12753401}{35035545600}$	$-\frac{6906257}{245248819200}$	$\frac{77}{88473600}$

stead of just the phonon vacuum:

$$H_{\text{eff}} = \langle\xi|\tilde{H}|\xi\rangle = J_0 N + \sum_i \sum_{n=1}^{7} J_n \vec{S}_i \cdot \vec{S}_{i+n} + \text{multi-spin terms} \tag{7}$$

This allows for a direct comparison with the flow equation result of Raas et al.[15], which is equivalent to a 2nd order expansion ($k = 2$) with phonon shift $\xi = -\langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle g \approx (1/4 - \ln 2)g \approx 0.44g$. Here the spin correlator is approximated by its value in the isotropic Heisenberg chain. For a comparison see the small squares and the thin dashed line in Figure 1.

In Table I we list the expansion coefficients of the resulting long-ranged exchange interactions J_n that contribute to our effective Hamiltonian Eq. (7). We neglect interactions that involve more than two spin operators. The phase transition line in Figure 1 is obtained by equating the effective frustration $\alpha_{\text{eff}} := J_2/J_1$ with the critical value α_c of the next-nearest-neighbor spin chain (see Eq. (4)), where for the phonon shift we use the "neutral" value $\xi = 0$, i.e., the vacuum. Except for a constant offset, the analytical result matches the DMRG data quite well. In particular, the slope of the critical line is captured correctly.

The convergence of our analytical approach is illustrated in Figure 3, depicting the phase transition lines obtained for increasing expansion order. We observe an oscillatory behavior, such that reliable data results only beyond order $k \geq 8$. For lower orders, α_{eff} may not even reach the critical value, as is evident from the order 6 data shown in the inset. Nevertheless, for order 6 the phase transition can be estimated using a Shanks transformation[24].

We also checked, if the phonon shift $\xi \approx 0.44g$ proposed by Raas et al.[15] leads to an improved effective description, but clearly the dot-dashed line in Figure 3 deviates qualitatively from the DMRG data. Within the flow equation approach this shift was motivated by the technical requirement for normal ordering such that the phonons couple to $(\vec{S}_i \cdot \vec{S}_{i+1} - \langle \vec{S}_i \cdot \vec{S}_{i+1}\rangle)$, but not by, e.g., some variational principle. Hence, the discrepancy is not too surprising. Interestingly, for $\omega \lesssim 3$ we obtain an almost perfect description of the phase transition, if instead we assume a phonon shift of $\xi \approx 0.44g/\omega$. As yet we did not find a reasonable physical motivation for this ω-dependence, and the good agreement might be accidental.

To summarize, using DMRG we obtained the, to date, most precise numerical result for the location of the quantum phase transition from the spin liquid to the dimerized phase in the one-dimensional Heisenberg model with local coupling to optical phonons. In addition, we proved the convergence of the unitary transformation approach that maps the full spin-phonon model to an effective frustrated spin model and allows an analytical calculation of the phase boundary in good agreement with the numerical data.

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